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Gord et al.

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(54) **SYSTEMS AND METHODS FOR HIGH-SPEED, SPECTROSCOPIC, GAS-PHASE THERMOMETRY**

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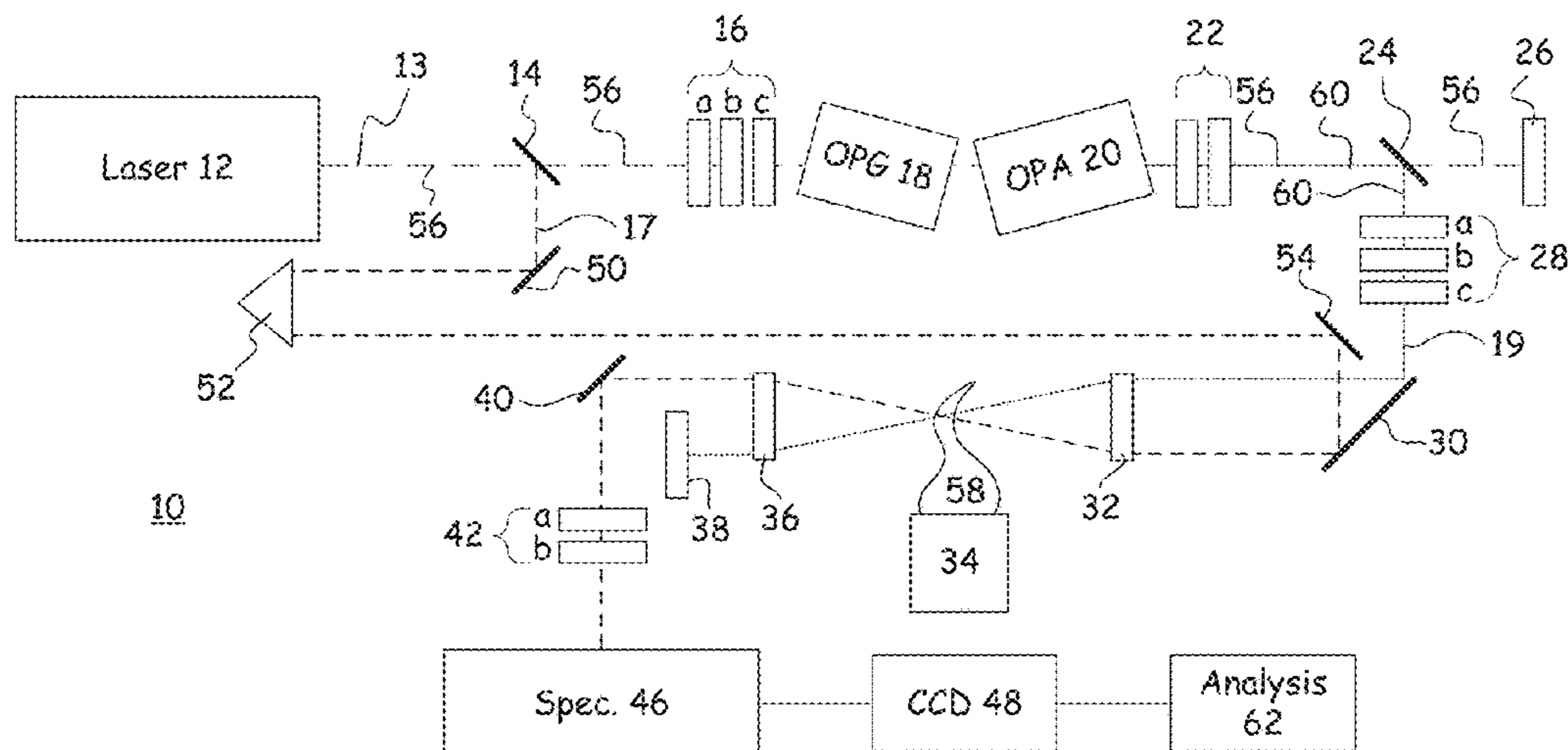
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(57) **ABSTRACT**

Systems and methods for measuring temperature in an environment by creating a first beam having an energy of about 50 mJ/pulse, and a pulse duration of about 100 ps. A second beam is also created, having an energy of about 2.3 mJ/pulse, and a pulse duration of about 58 ps. The first beam and the second beam are directed into a probe region, thereby expressing an optical output. Properties of the optical output are measured at a sampling rate of at least about 100 kHz, and temperature measurements are derived from the measured properties of the optical output. Such systems and methods can be used to measure temperature in environments exhibiting highly turbulent and transient flow dynamics.

1 Claim, 4 Drawing Sheets



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- continuation-in-part of application No. 15/582,792, filed on May 1, 2017, now abandoned.
- (60) Provisional application No. 62/404,397, filed on Oct. 5, 2016.
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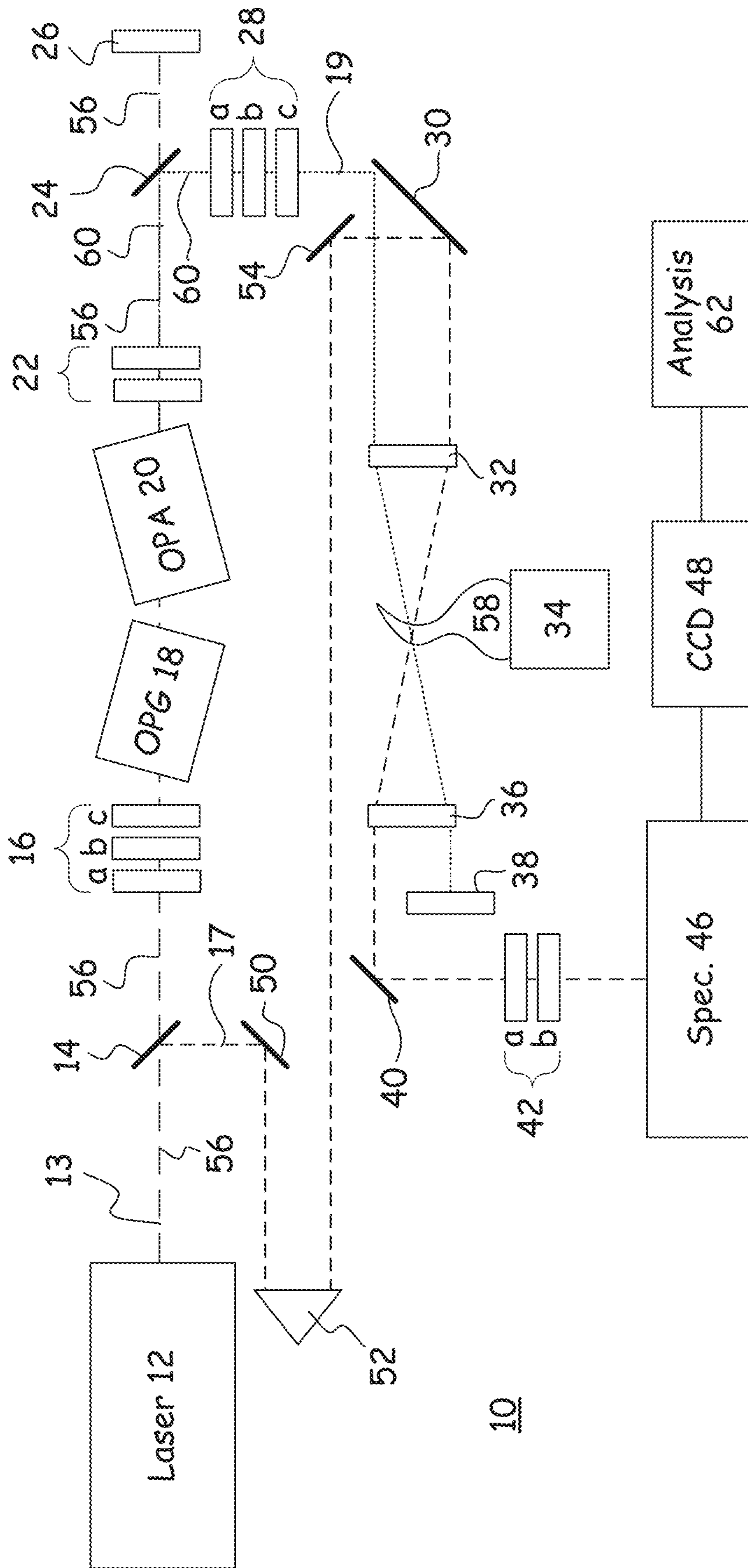
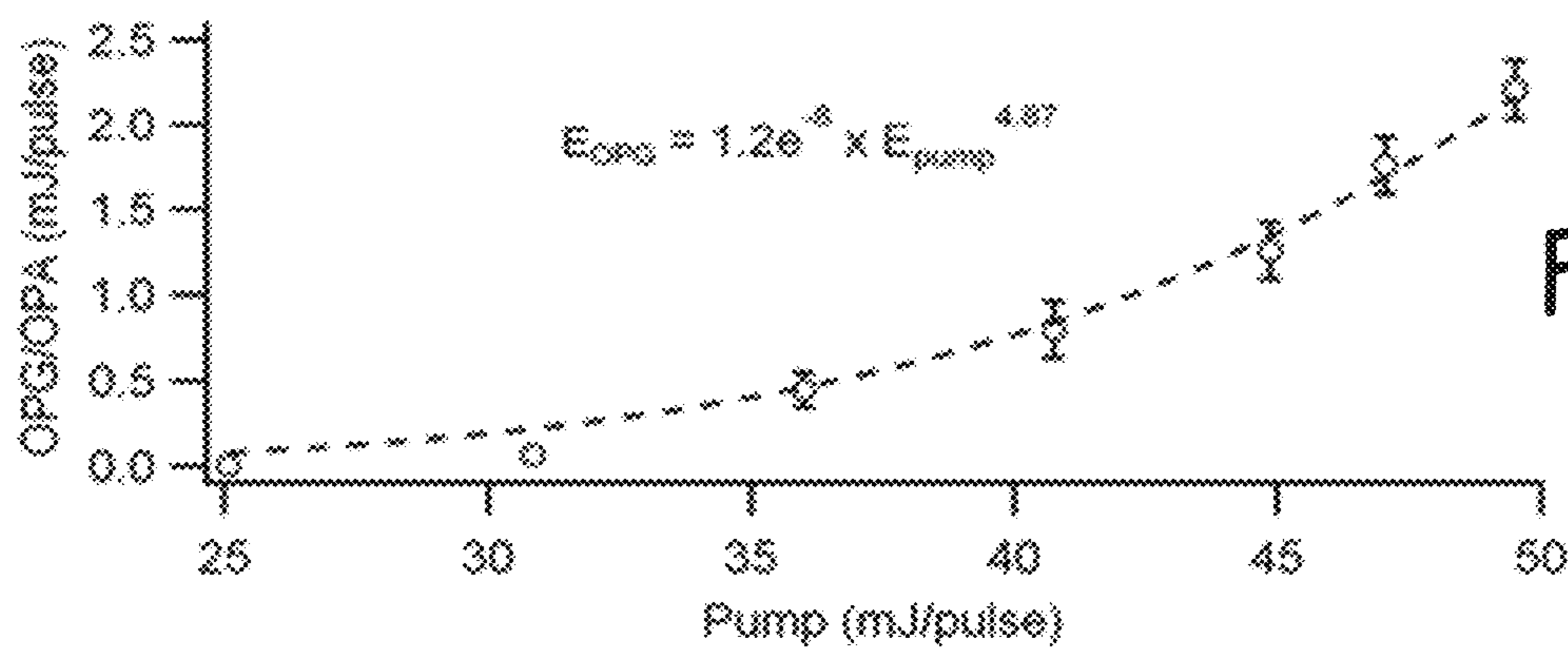
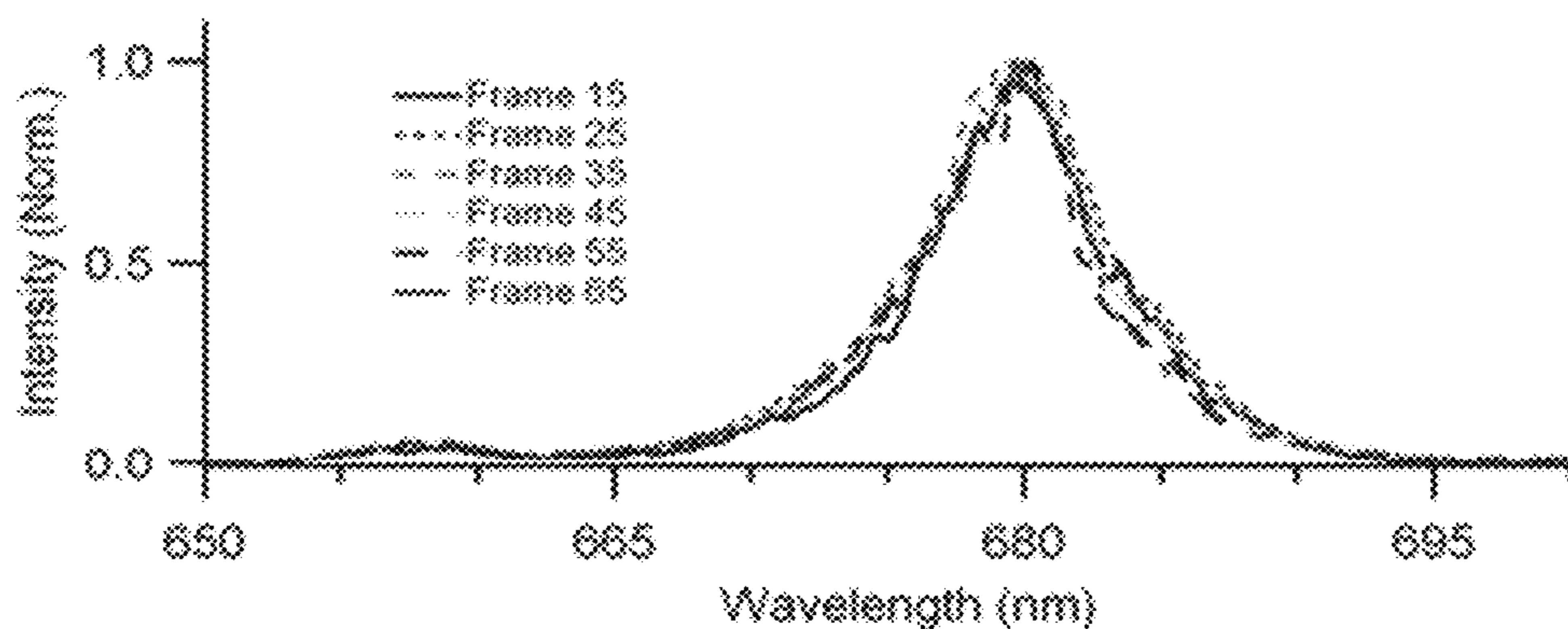
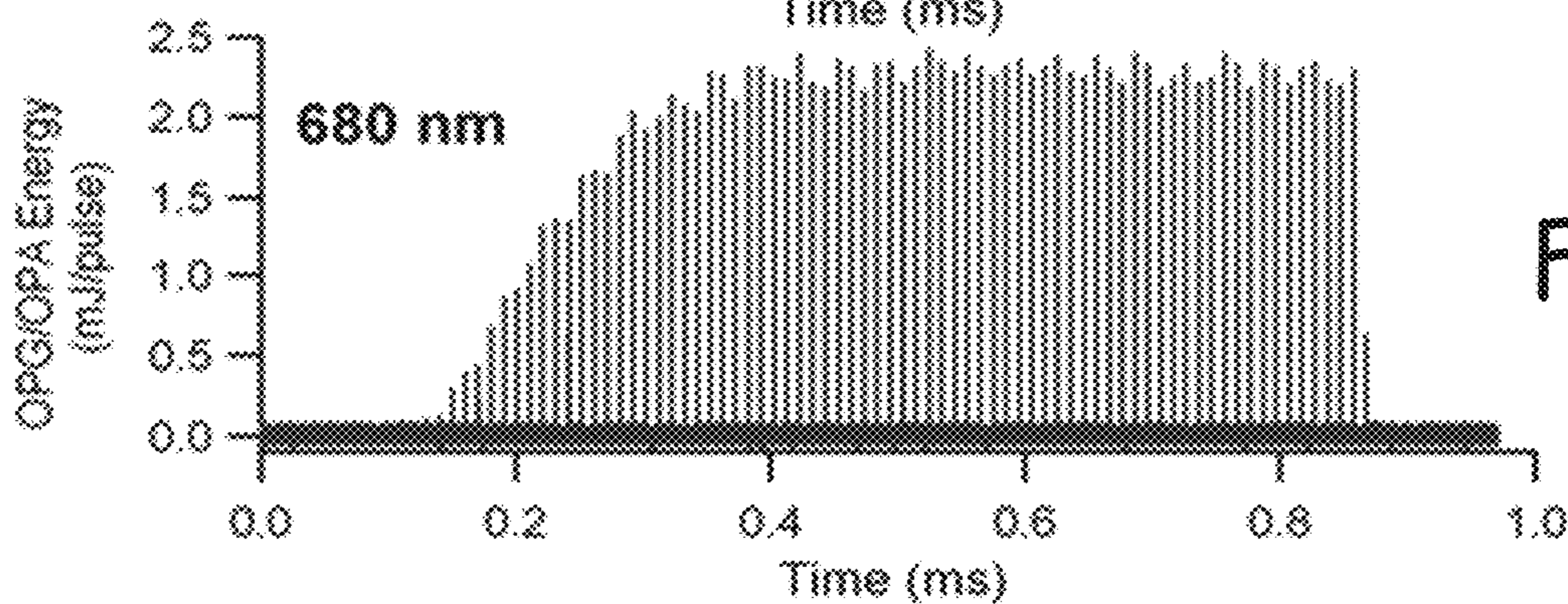
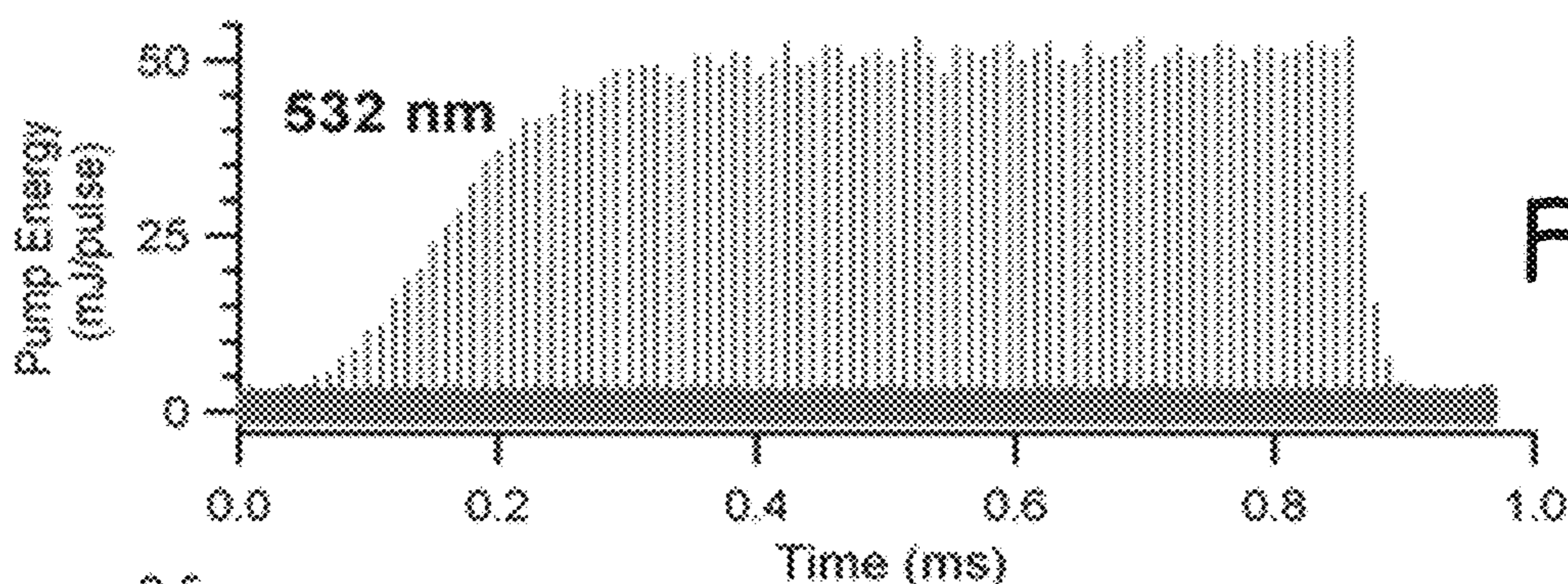


Fig. 1



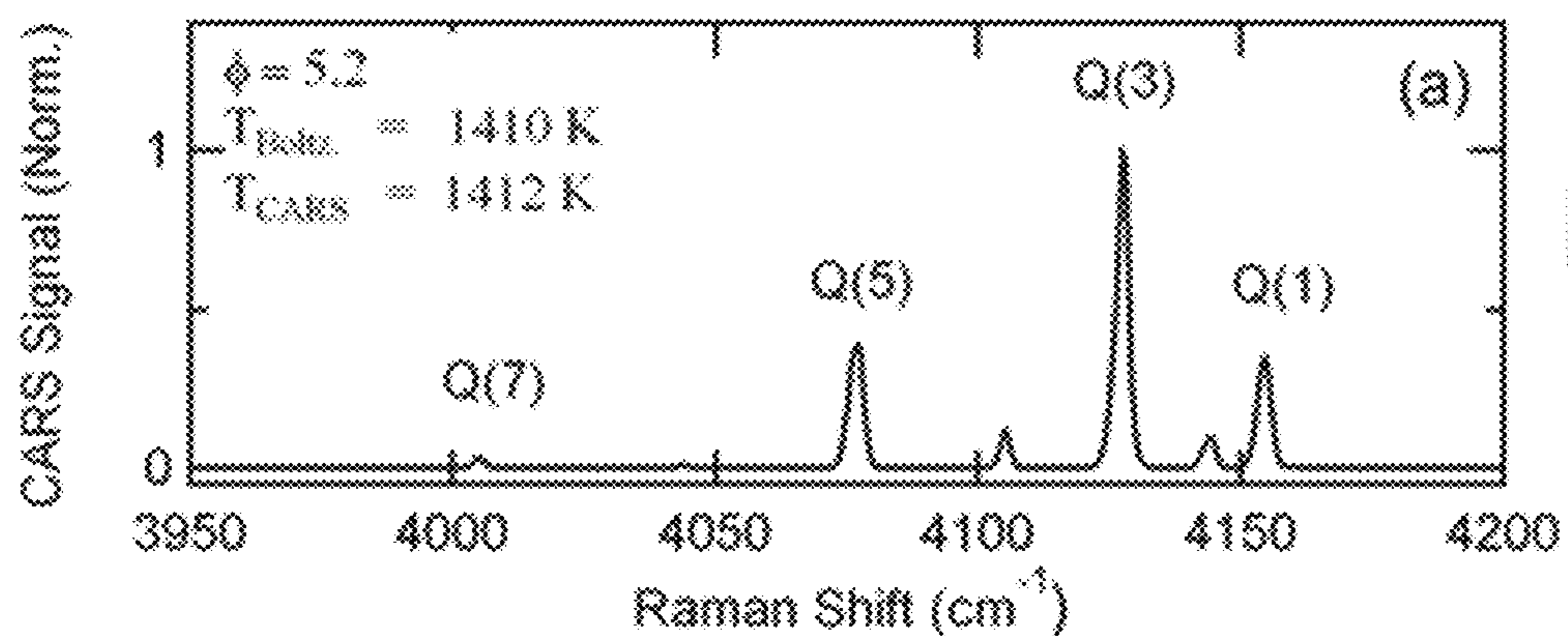


Fig. 3A

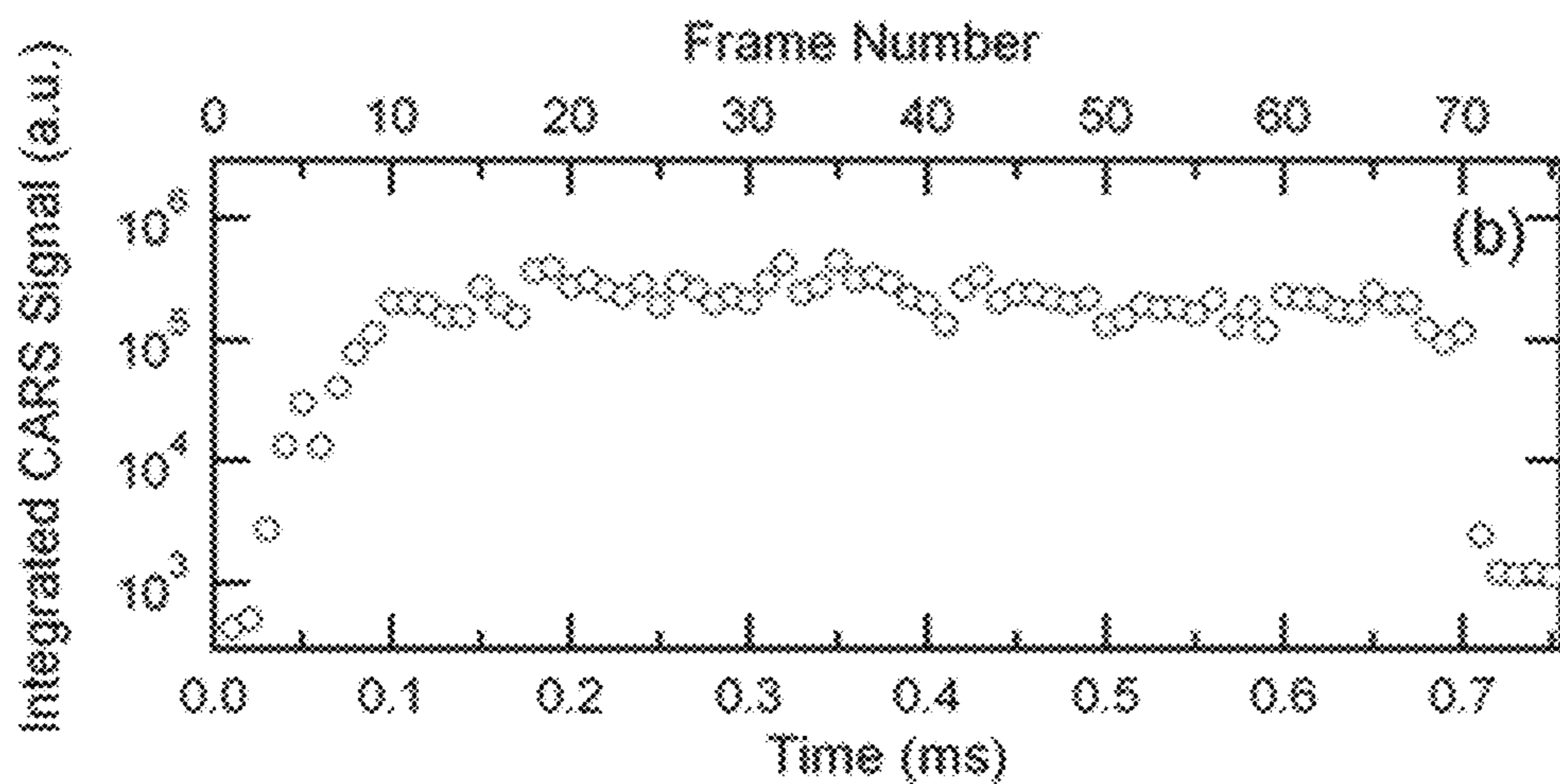


Fig. 3B

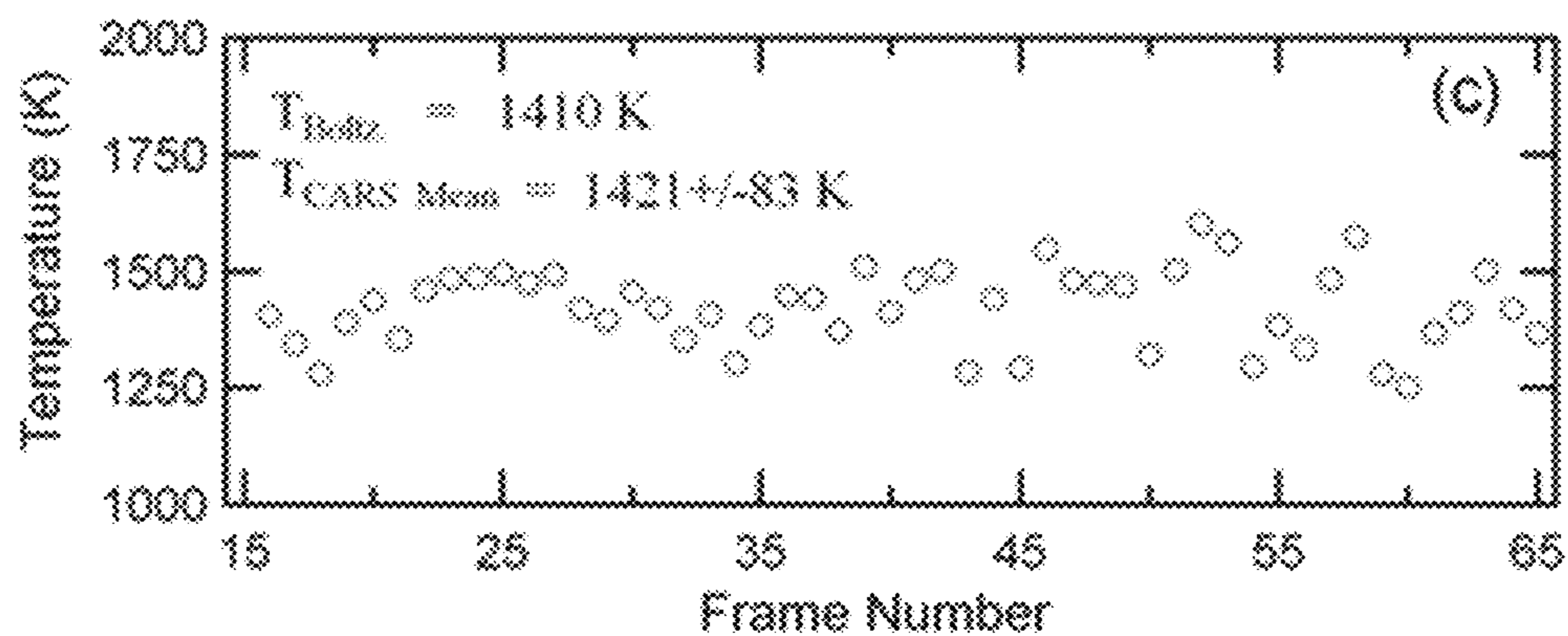


Fig. 3C

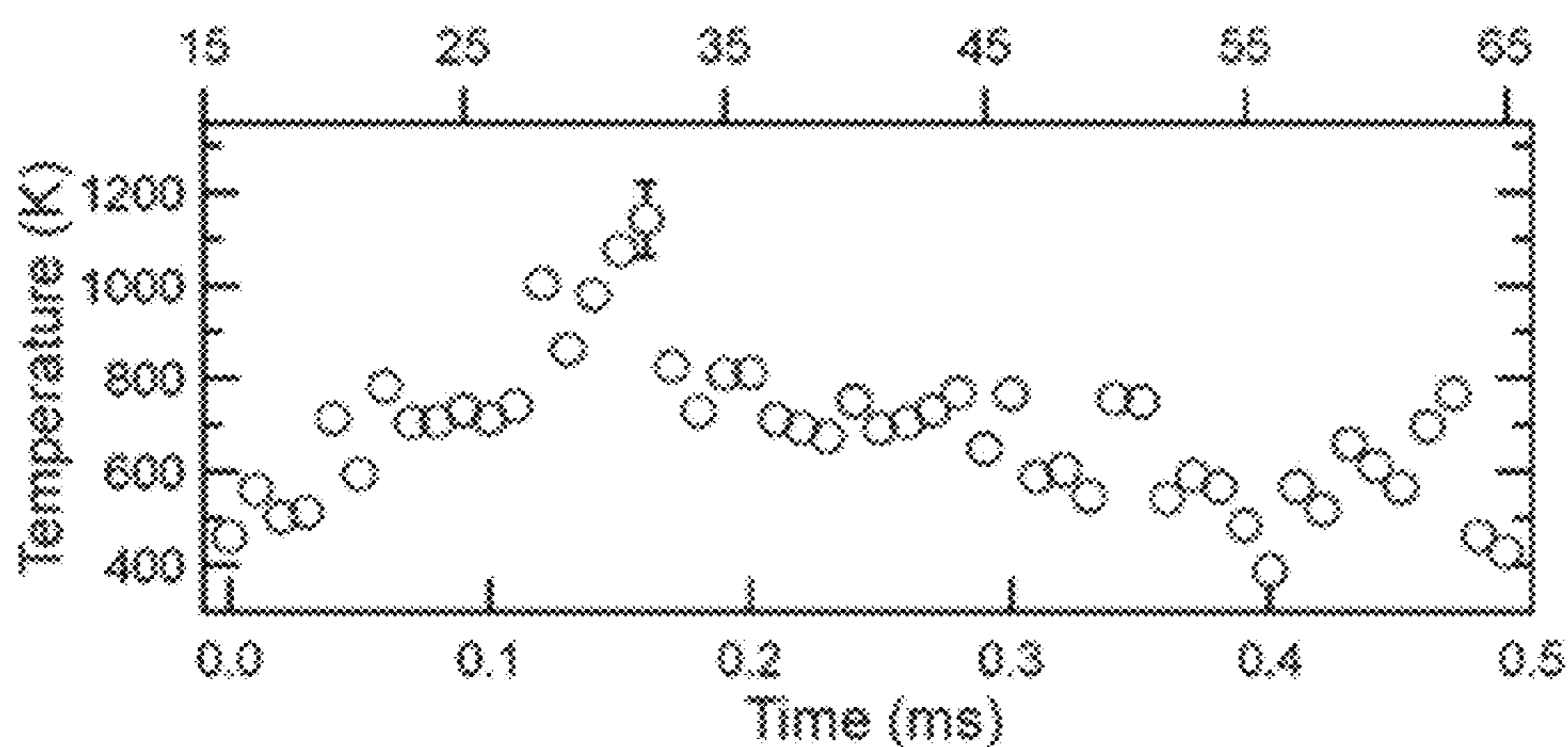


Fig. 4A

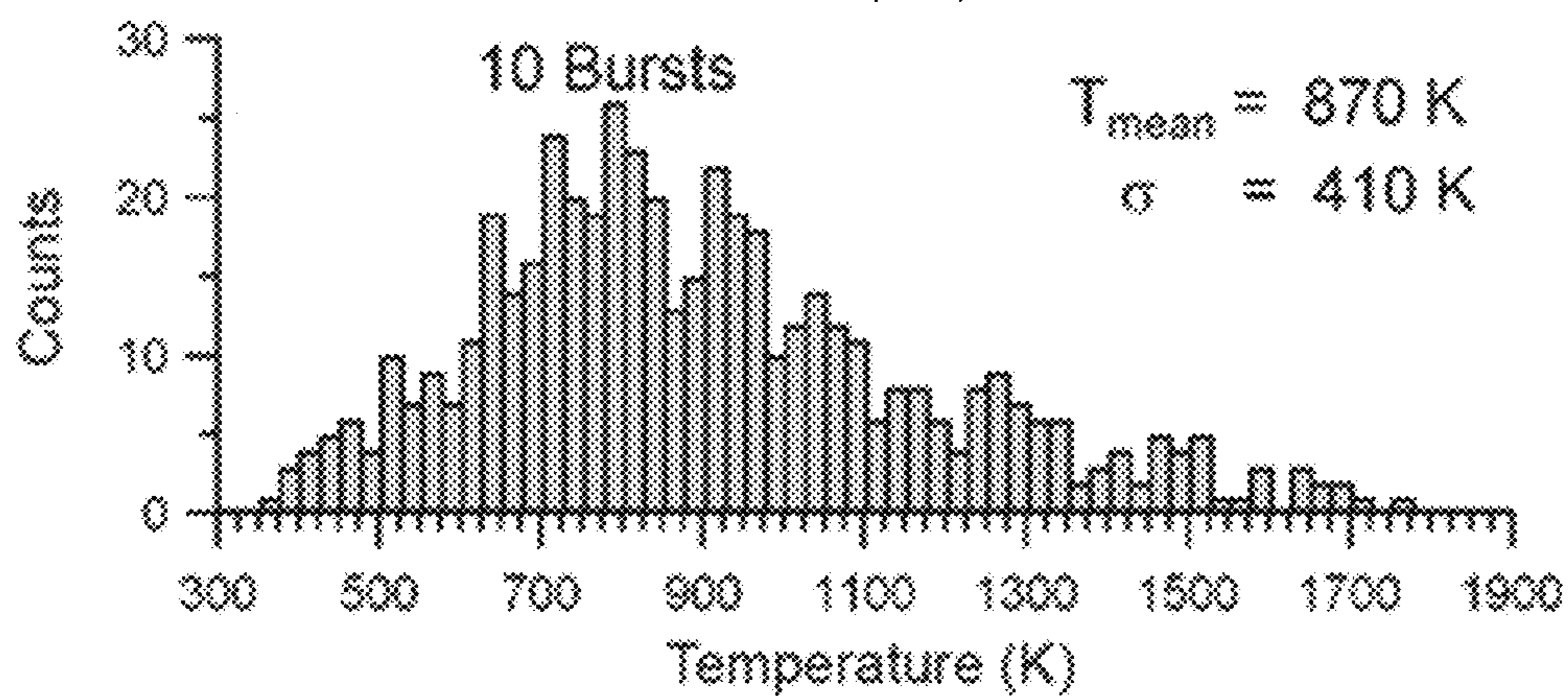


Fig. 4B

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**SYSTEMS AND METHODS FOR
HIGH-SPEED, SPECTROSCOPIC,
GAS-PHASE THERMOMETRY**

PRIORITY CLAIM

This application claims rights and priority on prior pending U.S. provisional patent application Ser. No. 62/404,397 filed 5 Oct. 2016, non-provisional patent application Ser. No. 15/582,792 filed 1 May 2017, and non-provisional patent application Ser. No. 16/052,763 filed 2 Aug. 2018, the entireties of which are incorporated herein by reference.

GOVERNMENT INTEREST

The invention described herein may be manufactured, used, and licensed by or for the Government of the United States for all governmental purposes without the payment of any royalty.

FIELD OF THE INVENTION

This invention relates to the field of temperature measurement. More particularly, this invention relates to coherent anti-Stokes Raman scattering (CARS) thermometry.

BACKGROUND OF THE INVENTION

CARS spectroscopy has proven to be a valuable tool for measuring temperature and major species concentrations in gas-phase reacting flows. CARS is a nonlinear diagnostic technique that relies on inducing Raman coherence in the target molecule(s) using two lasers. When this coherence is probed by a third laser, a coherent laser-like signal is generated in the phase-matching direction at a shifted frequency. This shifted signal is analyzed to achieve measurements of gas-phase temperature and species concentrations in reacting flows.

Over the last four decades, coherent anti-Stokes Raman scattering (CARS) spectroscopy has been widely used for thermometric and species-concentration measurements in gas-phase reacting and non-reacting flows. The spatio-temporally resolved nature of this approach, coupled with the coherent laser-like signal, allows for virtual time-freezing of the measured flow, thereby providing a near-instantaneous and highly accurate snapshot of the local temperature.

Until recently, gas-phase CARS-based temperature measurements were generally performed at rates of from about ten hertz to about twenty hertz. Other techniques have increased the measurements by more than two orders of magnitude to about five kilohertz, thereby enabling temporally-correlated, spatially-resolved measurements in moderately turbulent reacting flows with moderate Reynolds numbers (Re).

However, these measurement rates are not fast enough to investigate applications that exhibit highly turbulent and transient flow dynamics. Such applications include (1) high-Reynolds-number subsonic flows, (2) high-enthalpy hypervelocity environments in hypersonic propulsion, (3) high-speed transient thermometry for studying boundary-layer instability, (4) internal energy-exchange processes of interacting fluids, (5) shock-wave/boundary-layer interactions in high-speed flows, (6) explosion dynamics, and (7) flow dynamics of rocket propulsion.

For example, in high-enthalpy hypervelocity environments (application (2) above), measurements would preferably be performed at rates of about one-hundred kilohertz to

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about five megahertz (or from about twenty to about one thousand times faster than other techniques can provide).

What is needed, therefore, is a system that tends to address issues such as those described above, at least in part.

BRIEF SUMMARY OF THE INVENTION

The above and other needs are met by the systems and methods for measuring temperature as described herein. According to one embodiment, a method for measuring temperature is provided by creating a first beam having an energy of about 50 mJ/pulse, e.g. 50-200 mJ/pulse, and a pulse duration of about 100 ps, e.g. 50-200 ps. A second beam is also created, having an energy of about 2.3 mJ/pulse, e.g. 2-7 mJ/pulse, and a pulse duration of about 58 ps, e.g. 55-100 ps. The first beam and the second beam are directed into a probe region within an environment, thereby expressing an optical output from the environment. Properties of the optical output are measured at a sampling rate of at least about 100 kHz, e.g. 100-500 kHz, and temperature measurements are derived from the measured properties of the optical output.

Various embodiments of the current invention extend the measurement rate of state-of-the-art CARS-based thermometry by at least about twenty times by using a burst-mode laser architecture having a burst duration of from about one millisecond to about ten milliseconds and a characteristic pulse width of about 100 ps. The short duration pulse train of the burst mode laser is ideal for facilities where temporally correlated measurements must be performed within a very limited time window.

The developed technique and system allow for the enhancement of data-acquisition rate (kHz-MHz CARS), enable investigation of systems that exhibit highly turbulent and transient flow dynamics, and provide valuable data for model validation related to high-Reynolds-number subsonic flows, high-enthalpy hypervelocity environments for hypersonic-propulsion research, high-speed transient thermometry for studying boundary-layer instability and the internal energy-exchange processes of interacting fluids, shockwave/boundary-layer interactions in high-speed flows, explosion dynamics, and flow dynamics related to rocket propulsion.

In various embodiments according to this aspect of the invention, the first beam has a wavelength of about 532 nm, e.g. 532 nm \pm 1.0 nm. In some embodiments, the second beam has a wavelength of about 680 nm, e.g. 680 nm \pm 1.0 nm. In some embodiments, the first beam and the second beam have burst durations of at least about 1 ms, e.g. 1-10 ms. In some embodiments, the second beam is created by directing a portion of the first beam into an OPG/OPA. Some embodiments include an OPG/OPA of type-I beta-barium-borate crystals that are cut at an angle of about 21° (i.e. 21° \pm 0.1°) to an optical axis, with a crystal length of about 10 mm, e.g. 10-15 mm, and about an 8-10 mm² cross section. In some embodiments, the first laser is created by a Nd:YAG-based picosecond burst-mode laser. In some embodiments, the properties of the optical output are measured with a spectrometer. In some embodiments, the measurements are recorded with an electron multiplying charge coupled device camera. In some embodiments, the optical output is passed through a bandpass filter prior to the step of measuring the properties of the optical output. In some embodiments, the first beam is a combined pump and probe beam. In some embodiments, the second beam is a Stokes beam.

According to another aspect of the invention there is described a coherent anti-Stokes Raman scattering spectro-

raphy system, having a Nd:YAG-based picosecond burst-mode laser for producing a combination pump and probe beam having a wavelength of about 532 \pm 1.0 nm, an energy of about 50-200 mJ/pulse, e.g. 50 mJ/pulse, and a pulse duration of 50-200 ps, e.g. about 100 ps. An OPG/OPA formed of type-I beta-barium-borate crystals that are cut at an angle of 21 \pm 0.1 $^\circ$ to an optical axis, with a crystal length of 10-15 mm, e.g. about 10 mm, and an 8-10 mm² cross section, receives a portion of the combination pump and probe beam and thereby produces a Stokes beam having a wavelength of 680 \pm 1.0 nm, an energy of 2-7 mJ/pulse, e.g. about 2.3 mJ/pulse, a bandwidth of 150-250 cm⁻¹, e.g. about 200 cm⁻¹, and a pulse duration of 55-120 ps, e.g. about 58 ps. Optics direct the probe beam and the Stokes beam into a common probe region within an environment for a duration of 1-10 ms, e.g. about 1 ms, thereby expressing an optical output from the environment. A spectrometer receives the optical output and produces a spectrum.

A number of operational parameters have been presented as ranges of values as well as specific values within the defined ranges. The operation of the spectrography system described herein is fully operational within the defined ranges, as well as any subset of the defined ranges.

BRIEF DESCRIPTION OF THE DRAWINGS

Further advantages of the invention are apparent by reference to the detailed description when considered in conjunction with the figures, which are not to scale so as to more clearly show the details, wherein like reference numbers indicate like elements throughout the several views, and wherein:

FIG. 1 is a functional block diagram of a CARS system, according to an embodiment of the present invention.

FIG. 2A is graph of a burst profile for a pump beam with a one millisecond duration, a frequency of 100 kilohertz, and a wavelength of 532 nanometers, according to an embodiment of the present invention.

FIG. 2B is a graph of a burst profile for a probe beam that corresponds to the output of an OPG/OPA with a wavelength of 680 nanometers, according to an embodiment of the present invention.

FIG. 2C is a graph of the OPG/OPA beam spectra at 100 kilohertz, showing every tenth spectrum, according to an embodiment of the present invention.

FIG. 2D is a graph of the OPG/OPA output energy as a function of pump-pulse energy, according to an embodiment of the present invention.

FIG. 3A is a graph of a single-shot picosecond-CARS hydrogen spectrum recorded in an hydrogen/air flame that is stabilized over a Hencken burner at $\phi=5.2$, according to an embodiment of the present invention.

FIG. 3B is a graph of an integrated, single-shot, picosecond-CARS hydrogen signal as a function of time, according to an embodiment of the present invention.

FIG. 3C is a graph of a corresponding temperature for fifty consecutive laser shots, according to an embodiment of the present invention.

FIG. 4A is a graph of temperatures that have been extracted from a spectrograph of fifty consecutive single-shot, picosecond-CARS hydrogen spectra taken at 100 kilohertz from a turbulent flame, according to an embodiment of the invention.

FIG. 4B is a graph of temperature probability density functions (PDFs) for ten bursts (a total of 500 laser shots), according to an embodiment of the invention.

DETAILED DESCRIPTION OF THE INVENTION

Embodiments of the present invention enable coherent anti-Stokes Raman scattering (CARS) spectroscopy to be performed at kilohertz (kHz) rates through megahertz (MHz) rates, such as above about 100 kHz. CARS spectroscopy at these rates makes possible temperature measurements in environments exhibiting highly turbulent and transient flow dynamics, such as may be present in an environment, such as a combusting environment, for example. Typical CARS temperature accuracy is about plus or minus two percent.

Operational Overview

Generalized embodiments of the present invention can include (1) a burst-mode laser, (2) a broadband laser, and (3) a high-speed spectrally resolved CARS-signal detection system. In some embodiments, an OPG/OPA is implemented as the broadband laser, because of general limitations in the current state of alternate technologies.

With reference now to FIG. 1, there is depicted a CARS spectroscopy system 10 according an embodiment of the present invention. The system includes a burst-mode laser 12 for producing a beam 56 directed along a first path 13, defined as the path between the burst mode laser 12 and the beam dump 26. Burst-mode laser 12 is, in some embodiments, a high-energy, high-repetition-rate picosecond burst-mode laser, such as a Nd:YAG-based laser, that in some embodiments produces a generally green laser beam 56 with a diameter of about 6 mm and having an energy of about 50 mJ/pulse and a wavelength of about 532 nm. Beam 56 is, in some embodiments, nearly transform-limited with a full-width at half-maximum (FWHM) pulse duration of about 100 picoseconds (ps) and a measured linewidth of about 0.2 cm⁻¹.

The beam 56 encounters a beam splitter 14, which passes a portion of the beam 56 along the first path 13, and diverts another portion of the beam 56 along a second path 17, which is defined between the beam splitter 14 and the spectrometer 46. The portion of the beam 56 that is diverted along the second path 17 is, in some embodiments, used as a probe beam 56. The portion of the beam 56 that is passed through the beam splitter 14 is, in some embodiments, used to produce a Stokes beam 60, all as described in more detail hereafter.

The portion of the beam 56 that is passed by the beam splitter 14 continues along the first path 13 through conditioning optics 16, such as a half-wave plate 16a, a concave lens 16b, and convex lens 16c that in some embodiments reduce the beam 56 from about a 6 mm diameter to about a 3 mm diameter. The reduced and conditioned beam 56 then passes through a solid-state, broadband, wavelength-tunable device, which in some embodiments is an optical parametric generator/optical parametric amplifier (OPG/OPA) 18/20, which produces as an output the Stokes beam 60, which also passes along a portion of the first path 13.

Some embodiments of the OPG/OPA 18/20 use type-I beta-barium-borate (BBO) crystals that are cut at an angle of about 21 $^\circ$ to the optical axis. In some embodiments the crystal length is about ten millimeters, with about an 8-mm² cross section.

In the embodiment depicted, the burst-mode laser 12 produces high-energy, transform-limited picosecond laser pulses 56 at a repetition rate of about 100 kHz or higher, providing sufficient picosecond pulse energy to pump the OPG/OPA 18/20, and generate a high-energy broadband Stokes laser beam 60.

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In some embodiments, the Stokes beam **60** is a generally red beam with a wavelength of about 680 nm, a FWHM bandwidth of about 8 nm, a pulse duration of about 58 ps, a bandwidth of about 200 cm^{-1} , and a pulse energy of about 2.3 mJ.

In the illustrated embodiment, the Stokes beam **60** and residual portions of the beam **56** exit the OPG/OPA **18/20** and pass along the first path **13** through additional conditioning optics **22**, before encountering another beam splitter **24**. The beam splitter **24**, in some embodiments, allows the residual portions of the beam **56** to pass through along the first path **13** to a beam dump **26**. The beam splitter **24** diverts the Stokes beam **60** along a third path **19**, and through another set of conditioning optics **28**, such as a lens **28a**, bandpass filter **28b**, and half wave plate **28c**. After passing through the conditioning optics **28**, the Stokes beam **60** is folded by a mirror **30** to continue along the third path **19**.

Returning now to the probe beam **56** that was diverted by the beam splitter **14** along the second path **17**, it is delivered along the second path **17** to the folding mirror **30**. However, the second path **17** between the beam splitter **14** and the mirror **30** includes some delay elements, such as might include a longer path length, using mirrors **50** and **54**, and prism **52**, such that it leaves mirror **30** in phase with the Stokes beam **60**, which was delayed by the OPG/OPA **18/20**.

Some characteristics of the Stokes beam **60** are depicted in FIG. 2. Typical burst profiles showing individual pulse energy for the pump beam **56** (about 532 nm) and the corresponding Stokes beam **60** (in this case, a Stokes beam having a wavelength of about 680 nm) over about a one millisecond burst duration with a frequency of about 100-kHz are displayed in FIGS. 2A and 2B, respectively. The pulse-to-pulse standard deviation across the burst (after reaching a plateau) is about 4% and about 6% for the pump beam **56** and the Stokes beam **60**, respectively. The dynamic spectral profile for the Stokes beam **60** for a few pulses is displayed in FIG. 2C. The recorded pulse-to-pulse variation of the spectral width is about 5%. The dependence of the pulse energy of the Stokes beam **60** on the pulse energy of the pump beam **56** is depicted in FIG. 2D.

From a power-law fit it is observed that the intensity of the Stokes beam **60** is proportional to the pulse energy of the pump beam **56** raised to the power of about 4.87. In some embodiments the maximum parametric down-conversion efficiency reaches about 5%. The conversion efficiency is limited in some embodiments by the threshold of about 1.8 GW/cm^2 at which the coating of the OPG/OPA **18/20** is damaged. For burst durations that are longer than about one millisecond, a small deterioration of efficiency is observed in some embodiments, which might be attributed to heating of the OPG/OPA **18/20** and the resulting change of the phase-matching conditions.

The probe beam **56** and the Stokes beam **60** are directed onto and folded by the mirror **30** in such a manner that the second path **17** and the third path **19** are axial, one to another, after leaving the mirror **30**. The beams **56** and **60** then encounter, in some embodiments, a spherical lens **32** with a focal length of about 200 mm, at a relatively small incident angle of about 3° , to achieve a desirable phase-matching condition for all of the transitions covered by the bandwidth of the OPG/OPA **18/20**. In some embodiments, the spherical lens **32** crosses the probe beam **56** and the Stokes beam **60** at a measurement point within a probe region **58**, which is located within an environment where temperature is to be measured.

The interaction of the probe beam **56** and the Stokes beam **60** produces, in the embodiment described herein, a hydro-

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gen CARS signal that is indicative of, among other things, the temperature at the precise point in the probe region **58** as which the probe beam **56** and the Stokes beam **60** interacted, and at the exact time that the synched pulses of the probe beam **56** and the Stokes beam **60** interacted. This CARS signal is carried along the second path **17** with the probe beam **56**.

In the illustrated embodiment, probe region **58** may be a gaseous reaction region, such as a flame, emitting from a burner or engine **34**, where an additional electron that is characteristic of the temperature of the flame **58** is emitted along the path of the probe beam **56**. In some embodiments, the probe beam **56** (including the CARS signal) and the Stokes beam **60** are conditioned by lens **36** such that the second path **17** and the third path **19** are once again axial, one to another.

The Stokes beam **60** is then absorbed by a beam dump **38**, in some embodiments, and the probe beam **56** (with the CARS signal) is directed by mirror **40** through conditioning optics **42**, which in some embodiments include a focusing lens **42b** to focus the combined components of beam **56** into the spectrometer **46**. Conditioning optics **42** in some embodiments include an 8 nm bandpass filter **42b** centered at about 439 nm (such as Semrock, LD01-439/8-25.4), to minimize signal interference from flame emission and laser scatter.

The probe beam **56** and resulting hydrogen CARS signal is accepted by a high-speed spectrally resolved CARS-signal detection system **46**, such as a 0.25-m spectrometer **46** that is equipped with a 2,400 groove/mm grating. The output of the spectrometer **46** is observed by a recording device **48**, such as a high-speed EMCCD camera **48** (such as a Princeton Instruments ProEM-HS 1024BX3) with a 1024×1024 pixel array, which is used in some embodiments for the acquisition of the CARS spectra at a rate of about 100-kHz, and is phase-locked to be synchronized with the laser **12**. In this embodiment, the camera **48** chip is illuminated over about 10 pixels (which is equivalent to a frame-shifting time of about 8 microseconds) to achieve the signal-acquisition speed of about 100 kHz. The camera **48** chip size is about 13×13 microns. This high-speed EMCCD camera **48** can operate at up to about 333 kHz. For data-acquisition speeds greater than 333 kHz, the camera **48** could be replaced with a FASTCAM SA-Z (from Photron).

The output of the recording device **48** is input to an analyzer **62**, where the CARS signal is analyzed to determine properties of the environment **58**, such as temperature, according to known principles. The CARS signal is temperature dependent. The strength of the signal scales non-linearly with the difference in the ground state population and the vibrationally excited state population. Since the population of these states follows the temperature-dependent Boltzmann Distribution, the CARS signal carries an intrinsic temperature dependence as well.

In actual tests, single-laser-shot temperature measurements were performed in highly turbulent hydrogen/nitrogen jet-diffusion flames **58** at rates of 100 kHz. The Reynolds number for the jet flame **58**, based on a diameter of about 9.5 mm for the jet flame **58**, was about 10,000. The measurements were made at a distance of about 50 mm from the jet **34** exit. Information in regard to the measurements is discussed below, with reference to FIGS. 3 and 4.

Measurements were initially performed in an atmospheric-pressure, nearly adiabatic hydrogen/air flame **58** that was stabilized over a Hencken burner **34**. The burner **34** was operated at high flow rates to minimize heat losses to the burner **34**. The energies used for pump **56** and Stokes **60**

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pulses were about 2 mJ/pulse and about 2.3 mJ/pulse, respectively. FIG. 3A displays a single-shot spectrum for hydrogen at an equivalence ratio (ϕ) of 5.2. The Q-branch transitions of the $v'=1 \rightarrow v''=0$ vibrational band of hydrogen are shown in FIG. 3A. The extracted temperature $T_{CARS}=1410$ K agrees well with the adiabatic flame temperature $T_{adiab.}=1412$ K.

FIG. 3B displays typical integrated CARS signal as a function of time. The CARS signal becomes stable after the sixteenth pulse of the pulse train, and this signal level is maintained from Frame 15 to Frame 65 (50 measurement data points, which is equivalent to a measurement time of 0.5 ms). The corresponding temperature acquired from each single-shot spectrum is shown in FIG. 3C.

Depicted in FIG. 4A are temperature values derived from the hydrogen picosecond-CARS signal that was obtained in the turbulent flame during a one millisecond burst. A rapid temperature change in the turbulent flame can be observed. Probability-density functions (PDFs) of temperature derived from 500 single-shot spectra collected from ten bursts are depicted in FIG. 4B. The obtained mean temperature and standard deviation of the single-shot thermometry in turbulent flames 58 were 870 K and 410 K, respectively. The jet flame 58 has a very wide temperature distribution because of turbulent mixing of the fuel (hydrogen) and the oxidizer (ambient air).

The foregoing description of embodiments for this invention has been presented for purposes of illustration and description. It is not intended to be exhaustive or to limit the invention to the precise form disclosed. Obvious modifications or variations are possible in light of the above teachings. The embodiments are chosen and described in an effort to provide illustrations of the principles of the invention and

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its practical application, and to thereby enable one of ordinary skill in the art to utilize the invention in various embodiments and with various modifications as are suited to the particular use contemplated. All such modifications and variations are within the scope of the invention as determined by the appended claims when interpreted in accordance with the breadth to which they are fairly, legally, and equitably entitled.

The invention claimed is:

1. A coherent anti-Stokes Raman scattering spectrography system, comprising:
 - a burst-mode laser for producing a combined pump and probe beam having a wavelength of 532 ± 1 nm, an energy of 50-200 mJ/pulse, and a pulse duration of 50-200 ps,
 - an OPG/OPA for receiving a portion of the combined pump and probe beam and thereby producing a Stokes beam having a wavelength of 680 ± 1 nm, an energy of 2.3-2.7 mJ/pulse, a bandwidth of $150-250 \text{ cm}^{-1}$, and a pulse duration of 55-100 ps,
 - optics for directing the combined pump and probe beam and the Stokes beam into a probe region within an environment, thereby expressing an optical output from the environment,
 - a 0.25-m spectrometer having at least a 2,400 groove/mm grating for receiving the optical output and producing a spectra,
 - an electron multiplying charge coupled device camera for recording the spectra, and
 - electronics for converting the recorded spectra into temperature measurements.

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